

# Scaling for the Stark effect in the Rydberg atoms

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Equations determining the shifts and widths of the Rydberg states in a strong electric field (for an arbitrary atom) and the scaling ratios for near-threshold Stark resonances with  $n_1 \sim n \gg 1$ ,  $n_2$  and  $m \sim 1$  have been obtained. These scaling ratios have been confirmed experimentally.

1. The study of the Rydberg states of atoms and molecules has recently produced considerable interest. The resonances in the cross sections for photoionization of atoms in a static electric field  $\mathcal{E}$  at  $n = 15\text{--}40$  and  $E \approx 0$  have been observed experimentally.<sup>1–5</sup> Numerical calculations for hydrogen atoms<sup>6,7</sup> have shown that the positions and widths of these resonances coincide with the complex energies  $E^{(n_1, n_2, m)} = E_r - i\Gamma/2$  of the Stark quasisteady states. This situation makes it possible to verify experimentally the theory of the Stark effect in strong fields.

We have developed an analytic theory of the Rydberg states for an arbitrary atom [see Eqs. (2) and (3)]. Using  $1/n$  expansion, we obtained scaling ratios for the near-threshold resonances. These scaling ratios agree well with experiment and can be used for identification of the resonance quantum numbers.

We will use atomic units (unless specified otherwise);  $n = n_1 + n_2 + m + 1$  is the principal quantum number of the level, where  $n_1$ ,  $n_2$ , and  $m$  are the parabolic quantum numbers ( $m \geq 0$ ).

2. In calculating the energy of the states  $(n_1, n_2, m)$  with  $n \gg 1$  and  $m \ll n$ , we will

use the semiclassical quantization conditions with allowance for the corrections on the order of  $\hbar^2$  (Ref. 8), approximate separation of variables in the region  $r > r_a$ ,<sup>1)</sup> and the "hidden" symmetry of the Coulomb field.<sup>9</sup> Let  $\beta_{1,2}$  denote the separation constants, and  $\epsilon$  and  $F$  denote the reduced energy and the reduced external field

$$\begin{aligned} \epsilon &= 2n^2 E^{(n_1, n_2, m)} = \epsilon' - i\epsilon'' , \\ \epsilon'' &= n^2 \Gamma^{(n_1, n_2, m)}, \quad F = n^4 \mathcal{E} , \end{aligned} \quad (1)$$

where  $\Gamma^{(n_1, n_2, m)}(\mathcal{E})$  is the width of the  $(n_1, n_2, m)$  level which is associated with the ionization of the atom by the field  $\mathcal{E}$ . We can then determine  $\epsilon$ ,  $\beta_1$ , and  $\beta_2$  from the equations

$$\begin{aligned} \beta_1 (-\epsilon)^{-1/2} f(z_1) - \frac{F}{8n^2} (-\epsilon)^{-3/2} g(z_1) &= \nu_1 , \\ \beta_2 (-\epsilon)^{-1/2} f(z_2) + \frac{F}{8n^2} (-\epsilon)^{-3/2} g(z_2) &= \nu_2 , \\ \beta_1 + \beta_2 &= 1 , \end{aligned} \quad (2)$$

whose derivation will be discussed below. Here  $z_1 = -16\beta_1 F/\epsilon^2$ ,  $z_2 = 16\beta_2 F/\epsilon^2$ ,

$$\nu_i = \left(1 - \frac{\delta}{n}\right) \left(n_i + \frac{m+1}{2}\right) / n, \text{ where } i = 1 \text{ or } 2,$$

and  $\delta = \delta(n_1, n_2, m)$  is expressed in terms of the quantum defects<sup>10</sup>  $\delta_l$  for a free atom,

$$\delta(n_1, n_2, m) = \frac{1}{n} \sum_{l=m}^{n-1} \left( C_{J, M-m; lm}^{JM} \right)^2 (2l+1) \delta_l, \quad (3)$$

$J = (n-1)/2$ ,  $M = (n_1 - n_2 + m)/2$ , and  $f(z)$  and  $g(z)$  are expressed in terms of the hypergeometric function:  $f(z) = F(1/4, 3/4; 2; z)$ ,  $g(z) = 2/3 F(3/4, 5/4; 1; z) + 1/3 F(3/4, 5/4; 2; z)$ . The parameter  $\delta$  takes into account the difference between the atomic field and the Coulomb field (in the region  $r \lesssim r_a$ ). The presence of the Clebsch-Gordon coefficients in (3) is attributable to the hidden symmetry group of the hydrogen atom:  $\text{SO}(4) = \text{SO}(3) \circ \text{SO}(3)$  and  $\mathbf{L} = \mathbf{J}_1 + \mathbf{J}_2$ , where  $\mathbf{L}$  is the orbital angular momentum and  $\mathbf{J}_i$  are the generators of one of the  $\text{SO}(3)$  subgroups. Since  $\delta_l$  decreases rapidly with increasing  $l$  (Ref. 10), the sum in (3) actually retains several leading terms. Asymptotically  $\delta(n_1, n_2, m) \propto 1/n \rightarrow 0$  as  $n \rightarrow \infty$ , but at  $n \sim 30$  they are not yet small.<sup>2)</sup>

Since the corrections which were ignored in (2) are no greater than  $n^{-4}$  (for  $m=0$ ), system of equations (2) is accurate enough for the Rydberg atoms. In the limit  $\mathcal{E} \rightarrow 0$  solution (2) is in agreement with perturbation theory up to terms of order  $\mathcal{E}^3$  inclusively. Using (2), it is possible, however, to consider the case in which the field is strong (to within values which are comparable with the field at the electron orbit,  $n^4 \mathcal{E} \sim 1$ ).

Although Eq. (2) can be solved numerically, at  $n \gg 1$  it is reasonable to use the 1/

$n$  expansion. For the states with  $n_1 \sim n \gg 1$ ,  $n_2$  and  $m \sim 1$  we find

$$\epsilon_{n_1, n_2, m} = \epsilon_0 + \frac{p}{n} \epsilon_1 + \frac{1}{n^2} (p^2 \epsilon_2 + \xi_2 + m^2 \eta_2) + \dots, \quad (4)$$

where  $p = 2n_2 + m + 1$ . In the limit  $n \rightarrow \infty$  system (2) reduces to the equation

$$(-\epsilon)^{1/2} = F(1/4, 3/4; 2; -16F/\epsilon^2), \quad (5)$$

whose solution will be denoted in terms of  $\epsilon_{cl} \equiv \epsilon_0(F)$ . It is easy to show that  $\epsilon_{cl}$  increases monotonically along with  $F$ , crosses the boundary  $\epsilon = 0$  at  $F = F_* = 0.3834$ , and remains real for all  $F$  in the range  $0 < F < \infty$ . At  $F > F_*$  the next terms of the  $1/n$  expansion acquire an imaginary part. The terms of order  $1/n$  and  $1/n^2$  in (4) in this case are expressed in terms of  $\epsilon_{cl}(F)$  and its derivatives. Taking advantage of this situation, we find the scaling ratios

$$E_r^{(n_1, n_2, m)} = \frac{1}{2\tilde{n}^2} \epsilon_{cl}(\tilde{n}^4 \xi), \quad \Gamma(n_1, n_2, m) = \frac{p}{\tilde{n}^3} \left(1 - \frac{\delta}{n}\right) \gamma_{cl}(\tilde{n}^4 \xi), \quad (6)$$

where  $\tilde{n} = n_1 + (m + 1)/2 - \delta$ ,  $\gamma_{cl}(F) = \theta(F - F_*) (F(d/dF) - 1) \epsilon_{cl}^{3/2}$ , and  $E > 0$ . In the subthreshold region  $E < 0$  we have

$$E_r^{(n_1, n_2, m)} = \frac{1}{2\tilde{n}^2} \{ \epsilon_{cl}(\tilde{n}^4 \xi) + \eta((\tilde{n}/n_*)^2 \xi) - (\tilde{n}/n_*)^2 \eta(n_*^4 \xi) \}, \quad (7)$$

where  $n_* = \tilde{n} + p/2 = n - \delta$ , and  $\eta(F) = [-\epsilon_{cl}(F)]^{3/2}$ . Equations (6) and (7) have only one universal function  $\epsilon_{cl}(F)$ , which is determined from (5).

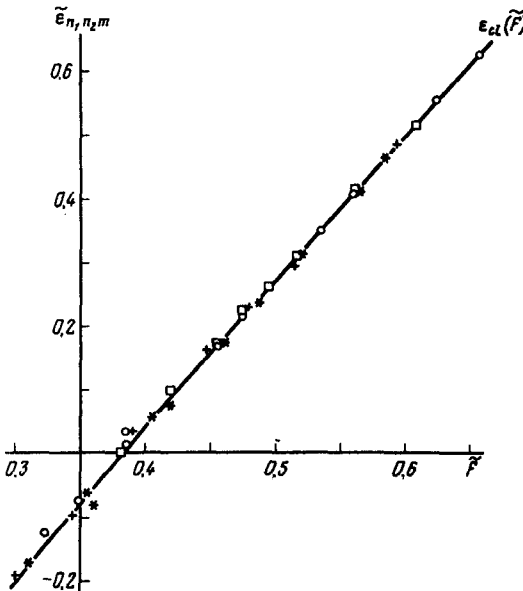


FIG. 1. Scaling for the above-threshold resonances. Solid curve  $\epsilon_{cl}(\tilde{F})$ ,  $\tilde{\epsilon}_{n_1, n_2, m} = 2\tilde{n}^2 E_r^{(n_1, n_2, m)}(\xi)$ ,  $\tilde{F} = \tilde{n}^4 \xi$ . The experimental points are explained in the text proper.

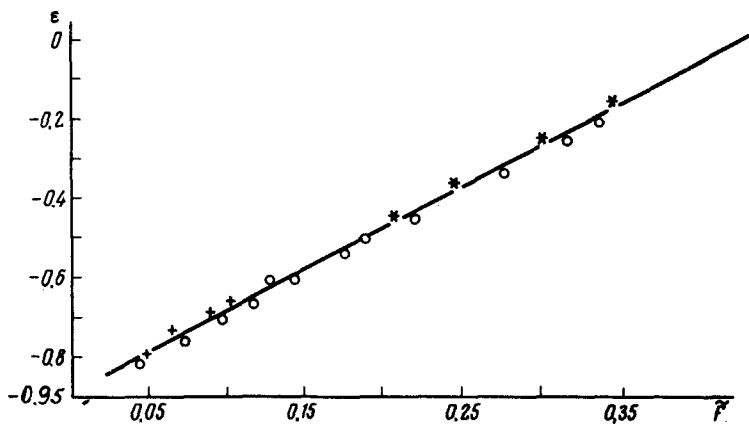


FIG. 2. Scaling (7) in the subthreshold region. The data for hydrogen (open circles) were taken from Refs. 4 and 5. The notation is otherwise the same as in Fig. 1.

3. Comparison with experiment. Satisfaction of scaling (6) for  $E_r^{(n,n,m)}$  is verified in Fig. 1. The experimental data points are:  $\circ$ —the  $(n_1, 0, 0)$  states of the hydrogen atom<sup>4</sup> for  $\mathcal{E} = 6.5$  and  $8.0$  kV/cm;  $\square$ —the  $(n_1, 0, 1)$  and  $(n_1, 1, 0)$  series in the hydrogen atom<sup>4</sup>;  $+$ —the data for rubidium<sup>1</sup> for  $\mathcal{E} = 2.189$  kV/cm (the four left points) and also for  $\mathcal{E} = 4.335$  and  $6.416$  kV/cm;  $*$ —the  $(n_1, 0, 0)$  states for sodium,<sup>2,3</sup>  $\mathcal{E} = 2.15$  and  $4.46$  kV/cm.

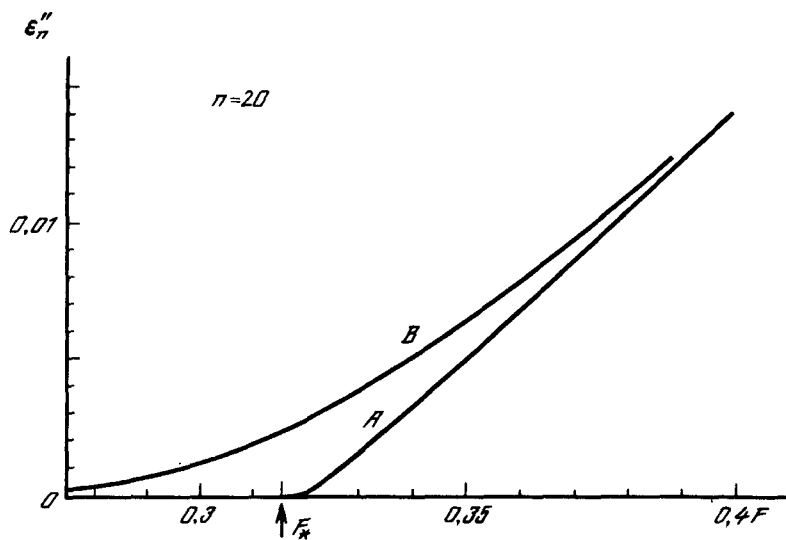


FIG. 3. Effect of barrier penetration on the calculation of the level width  $\epsilon''_n = n^2 \Gamma^{(n-1,0,0)}$ . Curve A—Solution of system (2) in the  $1/n$  approximation for  $\nu_2 = 1 - \nu_1 = 1/2n$  (here  $\epsilon''_n = 0$  for  $F < F_*$ ); curve B—solution of system (2) using replacement (8).

Satisfaction of (7) for the subthreshold resonances is illustrated in Fig. 2.<sup>1,2,4,5</sup> The number of experimental points in Figs. 1 and 2 could easily have been increased. In all cases we have considered the scaling ratios have been confirmed with good accuracy, both for hydrogen atoms and for other atoms.<sup>3)</sup>

With regard to the resonance widths, at  $F > 0.4$  the experimental points<sup>4</sup> conform well, according to (6), to the universal curve, but there is a divergence from scaling at lower values of  $F$ . Here the correction to the quantization due to the finite barrier penetrability should be taken into account. This procedure reduces to the substitution

$$\nu_2 \rightarrow \nu_2 - \frac{1}{2\pi n} \left\{ \frac{1}{2i} \ln \left[ \frac{\Gamma(\frac{1}{2} + ia)}{\Gamma(\frac{1}{2} - ia)(1 + e^{-2\pi a})} \right] - a \ln a + a \right\}, \quad (8)$$

where  $a = (1/\pi) \int_{\eta_1}^{\eta_2} |p_\eta| d\eta$  ( $\eta_1 < \eta < \eta_2$  is the subbarrier region). In the limit  $\mathcal{E} \rightarrow 0$  we can thus account for the well-known<sup>9</sup> threshold behavior of the widths  $\Gamma^{(n,n_3,m)}(\mathcal{E})$ . A numerical solution of system (2) with allowance for (8) gives a correct interpolation between the weak-field region and the scaling region  $F \gtrsim F_*$  (Fig. 3). We will compare the calculations with experimental data on the Stark resonance widths in a more detailed paper.

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<sup>1)</sup> Here  $r_a$  is the radius of the atomic core, which is assumed to be small in comparison with the mean radius of the Rydberg states,  $r \propto n^2$ .

<sup>2)</sup> In a rubidium atom, for example,  $\delta(n-1,0,0) = 0.768, 0.538,$  and  $0.414$ , when  $n = 20, 30,$  or  $40$ .

<sup>3)</sup> In the case of hydrogen, the quantum defects vanish. In other cases, it is important to take  $\delta(n_1, n_2, m)$  into account in Eqs. (6) and (7). To save space, we will omit here some details which are important for a correct interpretation of the experimental spectra for rubidium.<sup>1</sup>

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